Estimates of Acidic Deposition on Forest Areas

森林區酸沈降的估算

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ABSTRACT

In above cloud-base forests, pollutant/nutrient aerosols and gaseous species that are incorporating in clouds and fogs, are deposited on the forest canopy primarily through the interception of cloud/fog water (cloud deposition). This deposition mechanism is a hybrid between the wet and dry deposition mechanisms. To determine the relative importance of these deposition mechanisms in the forest areas, the database obtained from a field study which was conducted during the growing seasons of 1986-1988 at Mt. Mitchell, North Carolina, USA, was used. As a result, the deposition fluxes of sulfur (S) compounds were found primarily contributed through cloud capture mechanism (60 %) followed by incident precipitation (25 %) and dry deposition (15 %). As to the deposition fluxes of nitrogen (N) compounds, cloud, wet and dry deposition contributed about 50 %, 25 % and 25 %, respectively. A comparison of deposition estimates at Mt. Mitchell with those at other sites showed that the sulfate deposition at sites exceeding 1,200 m MSL in elevation in Bavaria (Germany) and eastern USA was almost identical within error limits. Reasons for large uncertainties in deposition estimates were discussed as also the mechanisms for redistribution of the deposited material on the forest canopy.

Key words: Forest decline, acidic deposition, wet deposition, dry deposition, and cloud deposition

摘　　要

在高於雲底的森林區，與雲霧結合的污染性／營養性的氣溶膠和氣態物質，主要是經由雲霧的受攔截(雲沈降)而沈降於其中。這種沈降機制是界於濕與乾沈降之間的混合型。在美國北卡羅萊納州 Mitchell 山所進行的實驗中，於 1986-1988 成長季節獲得的資料將被用來決定這些沈降機制在森林區的相對重要性。結果發現硫化物的沈降主要經由雲的攔截(百分之六十)、直接降水(百分之二十五)和乾沈降(百分之十五)。對氮化物的沈降而言，這三種機制的貢獻分別爲百分之五十、二十五和二十五。進一步與分佈於美東及德國巴伐利亞的測站資料比較，硫酸的沈降量則與在高度超過海拔1200公尺的測站所觀測的量相符。在本文中，也探討作沈降估算的不確定性與被沈降物質於森林中重新分配。

關鍵詞：森林死亡、酸沈降、濕沈降、乾沈降、雲沈降。

INTRODUCTION

In the past decade, the influence of acidic substances through deposition on forests, plants , and aquatic ecosystems have been widely debated. The SO_2 and NO_x gases which are mostly emitted by industries and through anthropogenic activities, are the major sources of the acidic deposition via long-range transport even over nonindustrialized areas. The assessments of wet and dry deposition have been quantitatively made by generalizing the data collected during long periods of time on a regional scale. In above cloud-base forests, pollutant/nutrient aerosols and gaseous species that are incorporating in clouds and fogs, are deposited on the forest canopy through impaction and sedimentation which are basically similar to dry deposition mechanisms. This deposition mechanism is therefore named as cloud deposition (Saxena and Lin 1990).

Recently, the forest decline at high elevation mountains in eastern USA has been suggested to be associated (NAPAP 1987) with the deposition of acidic substances on the forest canopy through dry, wet and cloud deposition pathways. The investigators (Saxena *et al.* 1989; Lin and Saxena 1991a, 1991b) have pointed out that the direct cloud capture mechanism by the forest (red spruce and Fraser fir) canopy of Mt. Mitchell was a major contributor to the deposition of acidic substances even though most hydrological input was received through direct precipitation. The acidic deposition due to direct cloud capture pathway assumes unusual importance in the mountain areas which are frequently immersed in clouds.

In this paper, the methodologies for estimating the acidic deposition on forested mountain area through the aforementioned three deposition pathways were introduced. The results from a field study conducted at Mt. Mitchell ($35^0$44'0 5"N, $82^0$17'15"W) State Park, North Carolina, USA during 1986-1988 growing seasons were ultilized to elucidate the relative importance of wet, dry and cloud deposition pathways to the acidic deposition on the forest areas. The deposition estimates were compared for those sites in North America and Europe and uncertainties in such estimates were also pointed out.

ESTIMATES OF ACIDIC DEPOSITION

2.1 Deposition Mechanisms and Methodology of Estimation

The removal process of particles and gases from the atmosphere to environmental surfaces is described as "deposition", which is important in delivering the airborne pollutants to the biosphere. The wet and dry deposition mechanisms are most often paid much attention. The former is the easiest to understand and measure. By way of precipitation, the pollutants scavenged by raindrops can be delivered to the environmental surfaces. Precipitation samples are easily collected and chemically analyzed. The precipitation intensity (I_p) was obtained in terms of precipitation amount collected within a projected time scale (e.g. mm h^{-1}). Thus, wet deposition flux (F_w) was computed as

$$F_w = I_p \cdot C_p, \qquad (1)$$

where C_p is the concentration of target species in the precipitation.

Unlike wet deposition, the mechanism of dry deposition is more complex and is not easily studied, because of its dependence on the aerodynamic characteristics material and the near-surface atmospheric boundary layer, the chemical and physical behaviors of the depositing material, and the structure and chemical nature of the collecting surface. The formula to estimate the dry deposition flux (F_d) was similar to F_w, expressed as

$$F_d = V_d \cdot C_d, \qquad (2)$$

where Vd is the dry deposition velocity which is defined as the ratio of the deposition flux divided by the airborne pollutant concentration (C_d) at some height above the surface layer. Direct methods for determination of Vd are subjected to the complexity mentioned above. Therefore, instead of direct measurements, inferential methods based on the gradient transport theory was developed to describe the mass transfer within the air-surface interface. Varying with atmosphere stability, deposition surface and properties of the depositing material, V_d, was

measured or inferred for particles and several gases. .

Aside from the above two mechanisms, cloud water deposition has been pointed out to be ecologically significant for atmosphere-canopy interactions (Lovett 1984), being responsible for significant hydrological inputs into forested ecosystems at high elevation mountain areas, which are frequently immersed in clouds formed due to the passage of large-scale systems or local topographically-induced circulations. This deposition mechanism is a hybrid between the wet and dry mechanisms since it behaves as in-cloud scavenging mechanism which is conceptually the prelude of precipitation scavenging, but the dominant mechanisms are impaction and sedimentation similar to those of dry deposition. As a consequence, cloud chemical deposition flux (F_c) due to direct cloud capture by the forest can be estimated by the production of the rate of cloud water deposition (I_c) which is similar to precipitation intensity, and the concentration (C_c) of target species in cloud water, expressed as

$$F_c = I_c \cdot C_c, \qquad (3)$$

Like Vd, at present, there is a lack of effective instruments to directly measure the rate of cloud water deposition.

2.2 Database and Models Used

In order to determine the relative importance of these deposition pathways, a field study conducted at Mt. Mitchell State Park, North Carolina, where the highest peak (2038 m MSL) in the eastern USA is located. The intensive observations began in May, 1986 and were performed through the entire growing seasons (from mid-May to the end of September) of 1986-1988. The meteorological paramters such as wind speed and direction, ambient temperature, relative humidity, barometric pressure, solar radiation, etc., were measured. The microphysical parameters such as cloud liquid water content and cloud/fog droplet siz distribution were also detected. In addition, the ambient air quality was monitored for O_3, SO_2 and NO_x gases using auto-analyzers. The particulate matters were also collected using filter packs. The precipitation data were obtained from a nearby NADP (National Atmospheric Deposition Program) site at Clingman's Peak (1,987 m MSL). The chemistry of collected precipitation and cloud/fog water were analyzed. The details regarding the experimental setup and measurements can be found elsewhere (Saxena and Lin 1990; Lin and Saxena 1991).

In order to estimate the acidic deposition due to direct cloud interception by the forest canopy, a cloud deposition model (CDM) (Lovett 1984; Saxena and Lin 1990) was used to compute the rate of cloud deposition. The ATDD (Atmospheric Turbulence and Diffusion Division, National Oceanic and Atmospheric Adminis tration) model (Hicks *et al.* 1987) was used for estimating the dry deposition rates of aerosol particles and gases. Besides, the wet depo-

sition was directly computed using the weekly NADP bulk precipitation samples. The detailed discussion of the methodologies regarding the estimation of wet, dry and cloud deposition has been given by Saxena and Lin (1990).

RESULTS AND DISCUSSION

As a result, Fig. 1 illustrates relative values with respect to the total mass fluxes for sulfur (S) and nitrogen (N) compounds. On the average, the cloud deposition did account for more than 60 % of S compounds and 50 % of N compounds, whereas, the wet deposition was responsible for 25 % of both compounds. Noticeably, dry deposition was more effective in transporting the N compounds compounds compared with the S compounds. As to the temporal variability, the acidic deposition for 1987 was apparently higher than those for other two years , primarily due to a significantly higher amount of precipitation and clouds intercepted by the forest in 1987. Notably, summers of 1986 and 1988 were recorded as climatologically drought years.

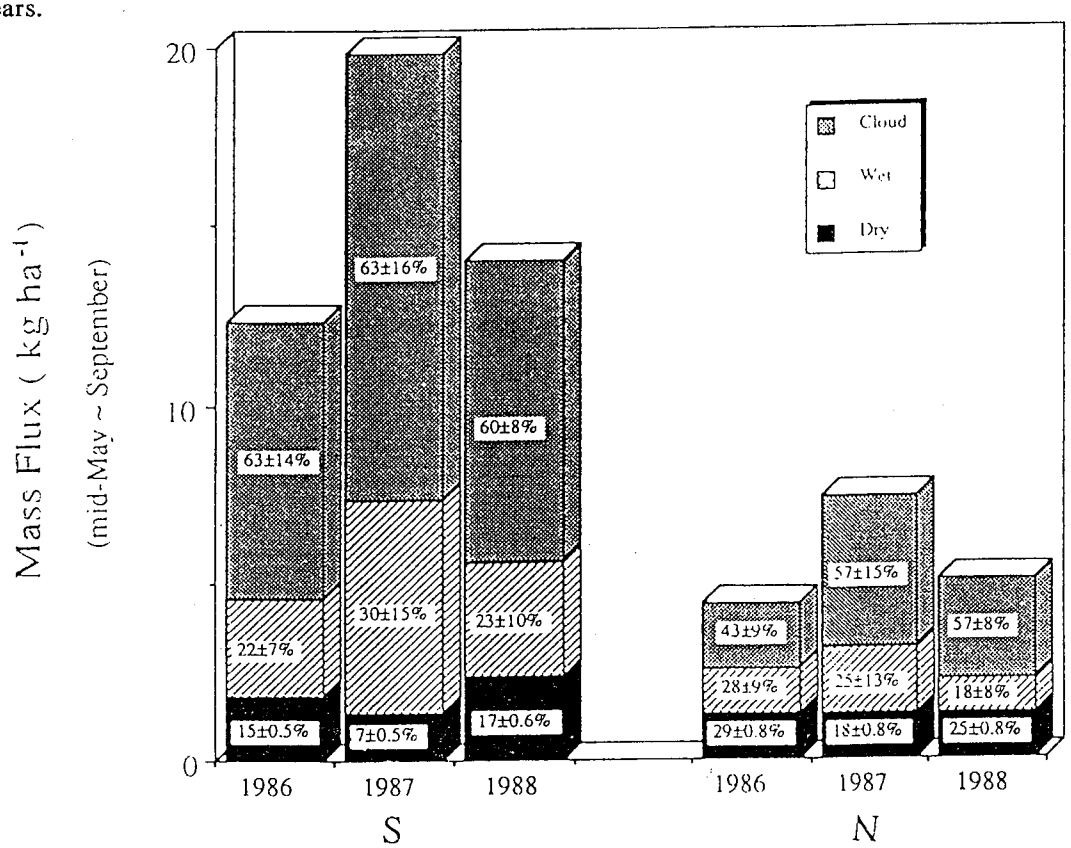

Figure 1. The contribution of dry, wet and cloud deposition pathways to the budget of sulfur (S) and nitrogen (N) compounds during the growing seasons of 1986-1988 in the Mt. Mitchell State Park, North Carolina, USA (after Lin and Saxena 1991b).

The determination of relative importance of wet and dry deposition is subject to some uncertainties mainly due to difficulties in obtaining the deposition velocities for the gases, however, the effect of varying deposition velocity on the flux may be of little consequence because of usually lower gaseous concentrations at remote sites. In the mountain areas which are frequently immersed in clouds, the cloud deposition is found to be more important in delivering the acidic substances to the forest canopy. The importance of this finding should be viewed together with the enhanced decline in the forest health at elevations exceeding 1,600 m MSL in the Mt. Mitchell State Park. The cloudbase is frequently observed (Saxena *et al.* 1989) at this altitude.

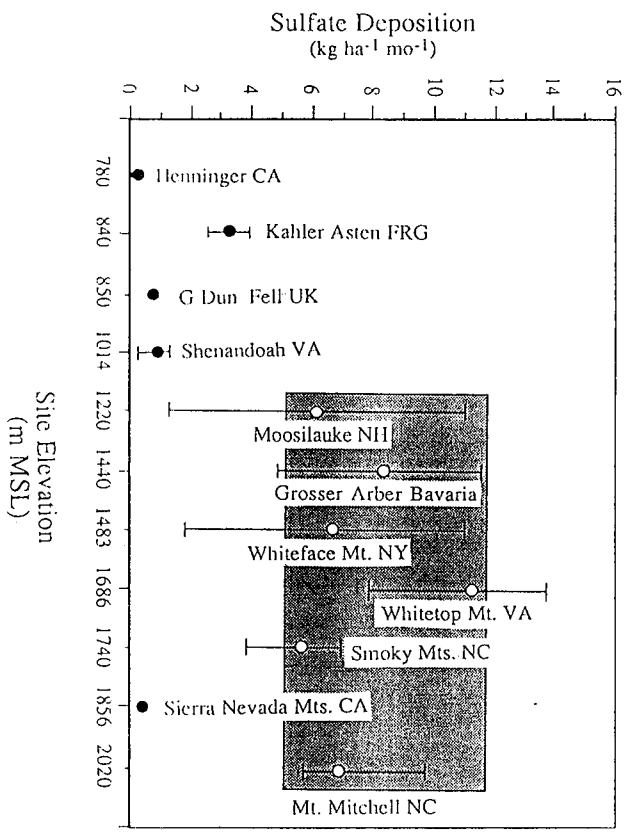

Figure 2. Plot of sulfate deposition as a function of elevation of various stations in Europe and North America. Open circles enclosed by the shaded area indicate those stations in USA and Germany for which the sulfate deposition is within a factor of two. The elevation of these sites exceeds 1,200 m MSL. Filled circles represent the sites with considerably less sulfate deposition (after Lin and Saxena 1991b).

In Fig. 2 the sulfate depositions for various sites in North A merica and Germany were plotted. Three methods for estimating the sulfate and water deposition on the forest canopy have been used: inferential deposition model, direct throughfall measurements, and educated guesses. Each of these methods was subjected to uncertainties arising from (1) the natural

variations in the forest canopy, (2) the natural variations in the cloud characteristics that the forest canopy intercepts, and (3) the natural variations in the wind speed coupled with inhomogeneous structure of the forest canopy resulting in the dependence of the cloud capture efficiency upon the type and location of the forest.

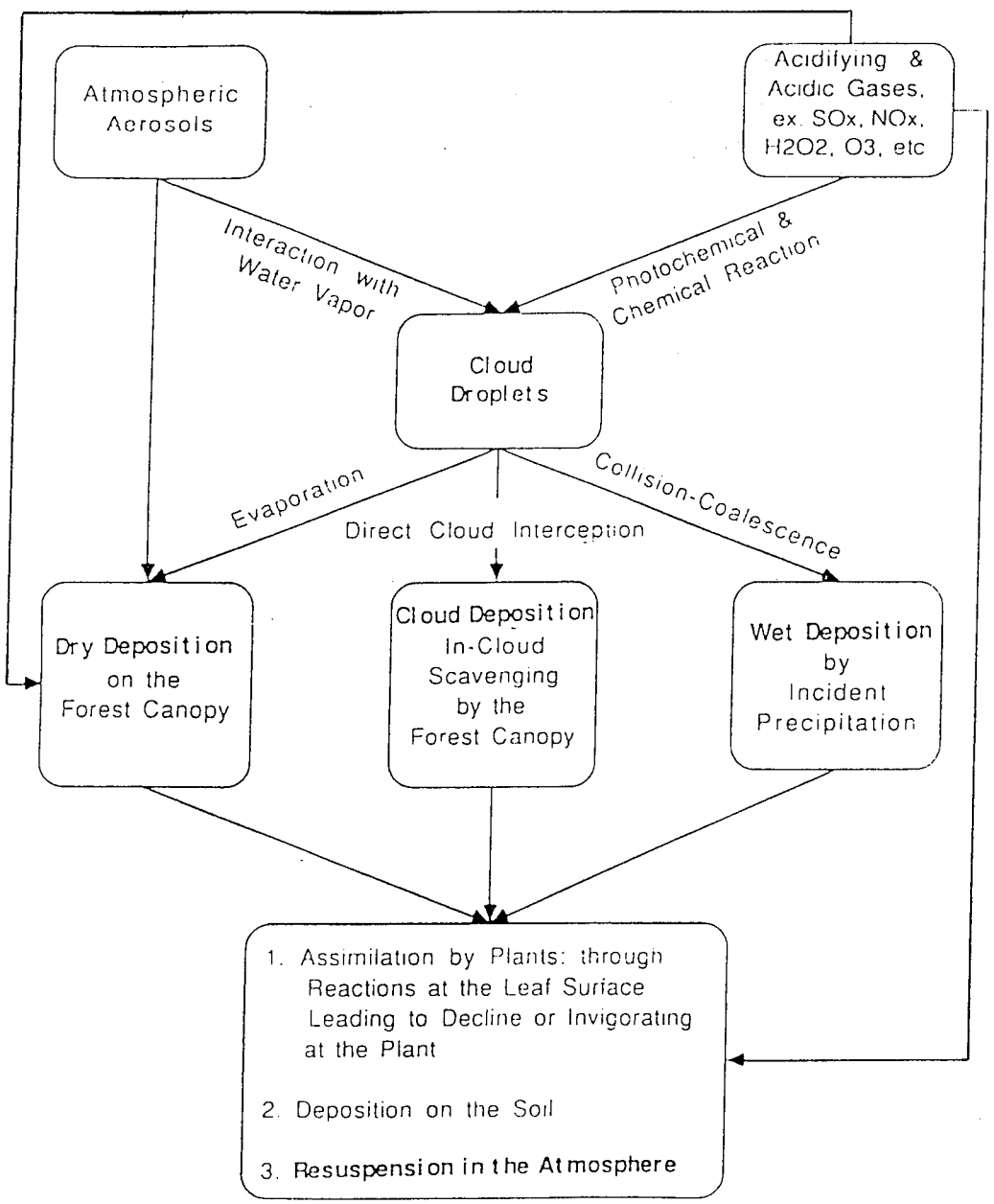
Atmospheric Aerosols
Acidifying & Acidic Gases, ex. SOx, NOx, H2O2, O3, etc
Interaction with Water Vapor
Photochemical & Chemical Reaction
Cloud Droplets
Evaporation
Direct Cloud Interception
Collision-Coalescence
Dry Deposition on the Forest Canopy
Cloud Deposition In-Cloud Scavenging by the Forest Canopy
Wet Deposition by Incident Precipitation
1. Assimilation by Plants: through Reactions at the Leaf Surface Leading to Decline or Invigorating at the Plant
2. Deposition on the Soil
3. Resuspension in the Atmosphere

Figure 3. Cloud droplets, being the chief sites for heterogeneous chemical reactions, play an important role in all the three modes of pollutant deposition, dry deposition, dry deposition, dry deposition, cloud deposition, and wet deposition. Due to windy environment at high elevations, the deposited material may also be resuspended in the atmosphere (after Lin and Saxena 1991b).

Two noteworthy features were evident from Fig. 2. First, at elevations exceeding 1,200 m MSL, the uncertainties in the sulfate deposition estimate were large, easily exceeding a factor of two. Secondly, within the bounds of errors, the sulfate deposition at five mountain sites in the eastern USA and one site in Bavaria (Germany) could be considered almost the same at site elevation exceedng 1,200 m MSL. The latter conclusion was significant in view of the fact that the forest decline was noticed at high elevations during the last decade.

Fig. 3 showed schematically how the three pathways of acidic deposition interacted with the clouds that were frequently encountered at high elevation sites. The deposited material on the forest canopy was redistributed by three mechanisms: assimilation by plants, deposition on the forest soil, and resuspension in the atmosphere. The first two mechanisms could lead to either decline or invigorating of the forest health depending upon the nature of the deposited material. The last mechanism was important in biosphere-geosphere interactions in as much as it changes the atmospheric contents especially over the high elevation forests and may have a wider ecological significance, hitherto unidentified. Its impact on cloud processes was already known.

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